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Improvement of GaN layer quality by using the bulk-GaN buffer structure grown by metalorganic chemical vapor deposition

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In this article, we propose a new buffer structure to obtain the high quality GaN epitaxial layers grown on sapphire substrates by a separate-flow reactor of metalorganic chemical vapor deposition (MOCVD). This buffer structure consists of 200–300 Å GaN nucleation layer/6 μm GaN-bulk layer. The bulk-GaN layers have also been prepared by MOCVD. The GaN epitaxial layer grown on this buffer structure exhibits a full width at half maximum (FWHM) of double-crystal x-ray diffraction of 170 arcsec, a FWHM of 300 K photoluminescence of 56 meV, an electron mobility of 400 $cm^2/V\,s$ at 300 K and 815 $cm^2/V\,s$ at 140 K, and a concentration of $3.6\times10^{17}\,cm^{-3}$ at 300 K. The GaN growth with this buffer structure has a wide growth window on the different nucleation-layer thicknesses. A good quality of GaN epitaxial layers can be obtained by using this buffer structure. © *1999 American Institute of Physics.* [S0021-8979(99)05118-X]

I. INTRODUCTION

GaN-based materials are emerging as the important materials for the optical devices in the blue and green wavelengths.[1–5] These devices are essential for full-color display and pickup head in high-definition digital-video-disk (HD-DVD) systems.[6] In addition, the wide direct band gap and excellent thermal conductivity make these materials good candidate for the high-temperature field-effect transistors (FETs).[7,8] However, a large lattice mismatch between GaN and sapphire substrate causes a large amount of dislocations embedded in the GaN epitaxial films.

In order to improve the quality of GaN epitaxial layer, some essential parameters of growth process and growth techniques have been investigated. These parameters of growth process consist of precursor flow, thickness, growth temperature, and V/III molar ratio of both GaN nucleation and epitaxial layers.[9,10] In addition, the growth techniques include the use of various single-crystal substrates[11–13] and the growth of a novel epitaxial laterally overgrown (ELOG) structure.[14] Recently, Nakamura *et al.* have fabricated the high-performance laser diodes with the cleaved mirror facets on bulk-GaN substrate.[15] This bulk-GaN substrate was grown by two-flow metalorganic chemical vapor deposition (MOCVD) and has a thickness of around 80 μm after removing the ELOG substrate. Tsuchiya *et al.* used the two-step growth method to obtain the thick cubic and hexagonal GaN epitaxial layer.[16] A thin GaN epitaxial film was first grown on GaAs substrate by gas-source molecular beam epitaxy and the wafer was then transferred to a hydride vapor phase epitaxy (HVPE) system to grow the thick GaN epitaxial layer. Detchprohm *et al.* also reported the two-step growth method to obtain a light-emitting diodes,[17] which is first to grow the thick GaN epitaxial layer on ZnO/sapphire substrate by HVPE and then was transferred to grow the Mg-doped GaN layer by MOCVD.

The purpose of this work is to propose a new approach of GaN buffer layer to improve the quality of GaN epitaxial layer. It consists of GaN nucleation layer / 6 μm GaN-bulk layer onto the sapphire substrate. It is found that the GaN epitaxial layer grown on this new buffer layer exhibits better electrical and optical characteristics than that grown on the conventional GaN nucleation layer.

II. EXPERIMENT

GaN films were grown in a separate-flow horizontal reactor by atmosphere-pressure MOCVD. The details of separate-flow horizontal reactor have been described in Ref. 18. The main reactant source was separated by a quartz plate, where we introduced the flow rate of 60 μmol/min in trimethylgallium (TMGa) and 5000 cc/min in NH_3 flowing into the upper and the bottom streams, respectively. In this work, the 4 μm GaN epitaxial layers were grown on the sapphire substrates through two structures of buffer layer: one is the conventional nucleation layer and the other is first to grow a 6 μm GaN bulk layer and then to grow the GaN nucleation layer, both grown on the sapphire substrates and denoted as structures A and B, respectively, as shown in Fig. 1. Prior to growth, the substrate was first precleaned at 1050 °C for 10 min in a hydrogen stream and then cooled down to 525 °C to grow the GaN nucleation layer with a thickness of 0–1000

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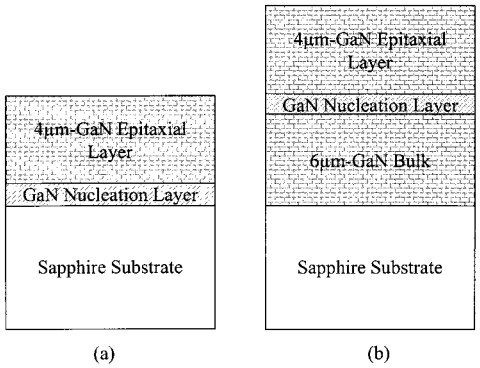

FIG. 1. Two buffer structures for GaN growth. Structure A is the conventional nucleation layer and structure B consists of nucleation layer/bulk-GaN layer, both grown on sapphire substrates.

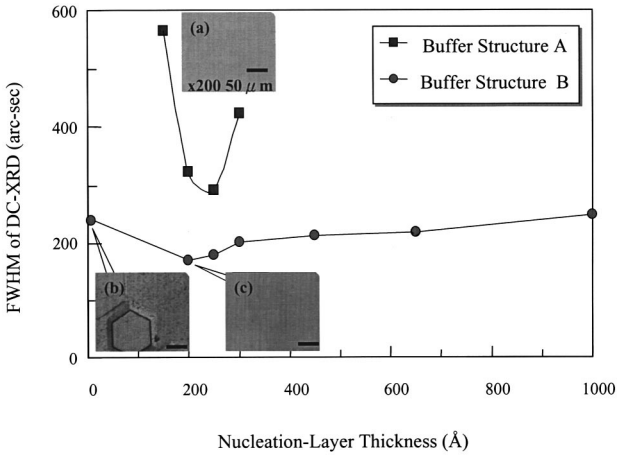

FIG. 2. FWHM of DC-XRD of GaN epitaxial layers as a function of nucleation-layer thickness for both buffer structures. The insets show the surface morphology of the GaN epitaxial layers with different nucleation-layer thicknesses for both structures.

Å, followed by raising to 1000 °C to grow the ~4 μm GaN epitaxial layer. For this studied approach, as shown in Fig. 1(b), the 6 μm bulk GaN layer was first grown on the sapphire substrate at 1000 °C. Sequentially, a nucleation layer and the ~4 μm GaN epitaxial layer were grown at high and low temperatures, respectively.

In order to identify the quality of GaN epitaxial films, the (0002) reflection of double-crystal x-ray diffraction (DC-XRD) was performed to analyze the crystallization using the (004) plane of Si as the first crystal. Photoluminescence (PL) was measured by an 18 mW/cm^{2}He–Cd($\lambda = 325$ nm) laser as the excitation light source with a spot size smaller than 0.2 mm through a focus lens and a narrow bandpass filter to block the laser harmonic lines. In addition, electron mobility and carrier concentration were measured in the temperature range of 77–300 K by van der Pauw method with a magnetic field of 5000 G and a current of 1 mA.

III. RESULTS AND DISCUSSION

If the 6 μm bulk GaN layer is directly grown on the sapphire substrate at 1000 °C, the bulk GaN layer will exhibit a surface morphology with hexagonal pyramids, electron mobility of 210 cm^2/V s, electron concentration of 1×10^{18} cm^{-3}, full width at half maximum (FWHM) of DC-XRD of 320 arcsec, and FWHM of 300 K PL of 75 meV. The poor-quality bulk GaN layer suffers from the large lattice mismatch between the sapphire and GaN epitaxial layer. Figure 2 shows the FWHM of DC-XRD as a function of nucleation-layer thickness for both the structures. For structure A, it has a FWHM of 565 arcsec at the nucleation layer of 150 Å, then decreases to a minimum of 290 arcsec at 250 Å thickness, and again becomes broader with further increasing thickness. For buffer structure B, however, the FWHM of DC-XRD can be lowered to the range of 290–170 arcsec at the nucleation-layer thickness of 0–1000 Å. Figure 2 also shows the surface morphology of GaN epitaxial layers with different nucleation layer thickness for both structures. The buffer structure A has a mirror-like surface morphology at the nucleation-layer thickness of 150–300 Å as shown in the inset (a) of Fig. 2. For the buffer structure B without nucleation layer, hexagonal pyramids will appear on the layer surface as shown in the inset (b) of Fig. 2, and the surface morphology becomes mirror-like with further increasing the nucleation-layer thickness in the range of 200–1000 Å [as shown in Fig. 2(c)]. It shows that buffer structure A has a narrow growth window in the nucleation-layer thickness due to a large lattice mismatch of 16% which existed between GaN film and sapphire. Akasaki *et al.* have successfully explained it by using a growth model with a nucleation layer.[19] It markedly influences the crystalline quality of GaN epitaxial films. The essential role of the nucleation layer in the GaN epitaxial grown includes the relaxation of strains, the reductions of microscopic fluctuations in crystalline orientation, free energy in the interface between GaN epitaxial layer and sapphire substrate, and original structural defects from the substrate. On the other hand, buffer structure B has a wider growth window in the nucleation-layer thickness to improve the FWHM of DC-XRD. Structure B also needs the growth of a nucleation layer on 6 μm bulk GaN layer because there are more strains embedded in 6 μm bulk GaN layer, even for thicker GaN films.[20,21]

For the 300 K PL spectra, all the GaN samples with buffer structures A and B exhibit a peak wavelength at 362 nm (3.425 eV), which is due to the near-band-edge emission. Figure 3 shows the FWHM of 300 K PL spectra of GaN epitaxial layers as a function of nucleation-layer thickness for both buffer structures. For the GaN growth on buffer structure A, the FWHM only exhibits a minimum of 66 meV at a nucleation layer thickness of 200–250 Å. Besides this range, it becomes broader again. However, the GaN layer with structure B has FWHM of 75 meV without depositing the nucleation layer, lowers to 65 meV at a nucleation-layer thickness of 200 Å, and then improves to 56 meV with further increasing the nucleation layer thickness beyond 300 Å. On the other hand, both buffer structures have a broader FWHM for a thinner nucleation layer than a thicker one. It is attributed to more strains embedded in GaN epitaxial layer and thus lead to a poor quality of the 4 μm GaN epitaxial layer. For the thickness of nucleation layer increasing from 200 to 1000 Å, the FWHM is almost kept at a constant of 56 meV, indicating that a lower shallow-level impurity exists in

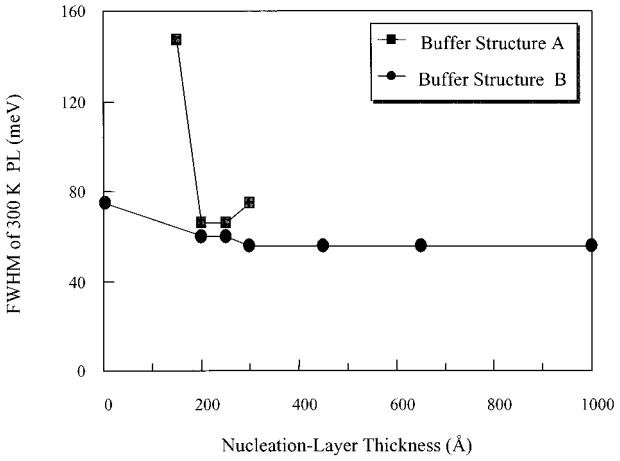

FIG. 3. FWHM of 300 K PL of GaN epitaxial layers as a function of nucleation-layer thickness for both buffer structures.

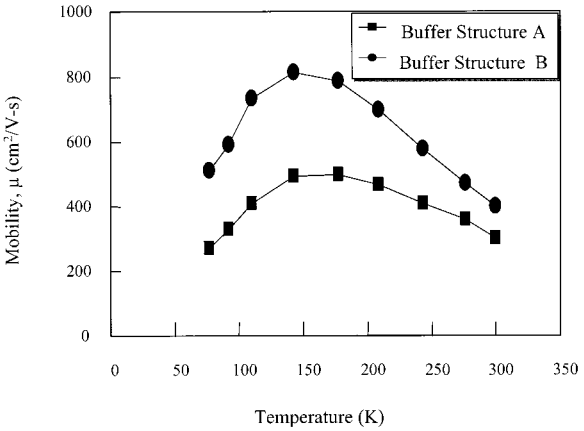

FIG. 5. Hall mobility as a function of temperature for the GaN epitaxial layers with a nucleation-layer thickness of 300 Å for both buffer structures.

the GaN epitaxial layer with buffer structure B.

The undoped GaN layers grown on both buffer structures always give an *n*-type conduction. Figure 4 shows the 300 K electron mobility and concentration as a function of nucleation-layer thickness for the 4 μm GaN epitaxial layers grown on both buffer structures. For buffer structure A, the mobility increases from 160 to 320 $cm^2/V\,s$ as the nucleation-layer thickness increases from 150 to 250 Å. With further increasing the nucleation-layer thickness, the mobility falls down. Meanwhile, the concentration lowers to a minimum of $6\times10^{17}\,cm^{-3}$ when the nucleation-layer thickness increases to 300 Å and then drops off for thicker thickness of nucleation layer. While the GaN epitaxial layers with buffer structure B have a similar trend of electrical properties to those with buffer structure A, the buffer structure B exhibits a maximum mobility of 400 $cm^2/V\,s$ and a minimum concentration of $3.6\times10^{17}\,cm^{-3}$ at the nucleation-layer thickness of 200 Å. As described above, good quality of GaN epitaxial layers can be obtained by using the buffer structure B. Figure 5 shows the electron mobility as a function of temperature in the range of 77–300 K for the GaN epitaxial layers grown on both buffer structures with a nucleation-layer thickness of 250 Å. When the temperature is lowered from 300 to 77 K, the mobility first increases, arrives to a maximum of 815 and 500 $cm^2/V\,s$ for both buffer structures A and B at 140 K, respectively, and then decreases below the temperature of 140 K. As well known, the mobility is dominated by the lattice and impurity scatterings at the temperatures above and below 140 K, respectively. As shown in Figs. 4 and 5, the electrical properties of GaN epitaxial layers can be improved by using buffer structure B with a nucleation layer thickness of 250–300 Å due to the significant reduction of defects and dislocations in the GaN films. These results can be compared to those of some researchers in the growth by using several alternative structures of double nucleation layer.[22–24]

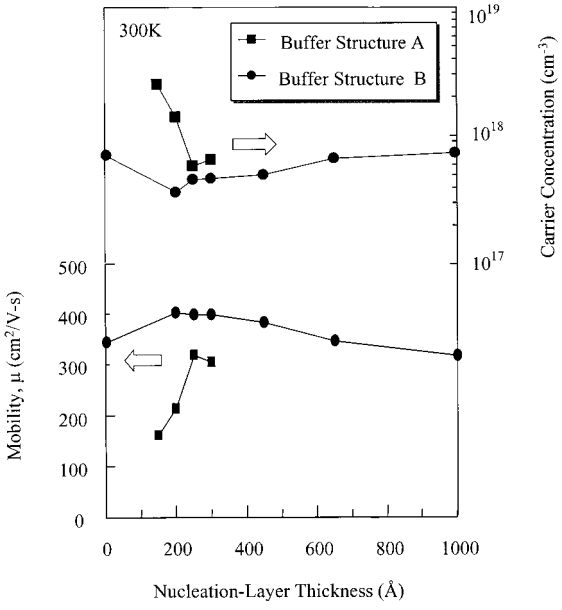

FIG. 4. Hall mobility and carrier concentration of GaN epitaxial layers at 300 K as a function of nucleation-layer thickness for both buffer structures.

IV. CONCLUSIONS

We have demonstrated a new approach of buffer structure with an optimal thickness of nucleation layer to improve the quality of GaN epitaxial layers grown by MOCVD. The optimal buffer structure consists of 200–300 Å nucleation layer/6 μm GaN epitaxial layer grown on the sapphire substrate. The GaN epitaxial layer grown on this buffer structure exhibits a FWHM of DC-XRD of 170 arcsec, a FWHM of 300 K PL of 56 meV, an electron mobility of 400 $cm^2/V\,s$ at 300 K and 815 $cm^2/V\,s$ at 140 K, and a concentration of $3.6\times10^{17}\,cm^{-3}$ at 300 K and $1.8\times10^{17}\,cm^{-3}$ at 140 K. The layer quality can be effectively improved by using the buffer structure B as compared to those using several alternative structures of double nucleation layer for the GaN growth. It is attributed to the effective relaxation of strains, the significant reductions of microscopic fluctuations in crystalline orientation, free energy in the interface between GaN epitaxial layer and sapphire substrate, and originated structural defects from the substrate.

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